

FORM PTO-1390 (Modified) (REV 11-98)		U.S. DEPARTMENT OF COMMERCE PATENT AND TRADEMARK OFFICE		ATTORNEY'S DOCKET NUMBER <b>TIL0003</b>	
TRANSMITTAL LETTER TO THE UNITED STATES DESIGNATED/ELECTED OFFICE (DO/EO/US) CONCERNING A FILING UNDER 35 U.S.C. 371				U.S. APPLICATION NO. (IF KNOWN, SEE 37 CFR) <b>09/869789</b>	
INTERNATIONAL APPLICATION NO. <b>PCT/DE00/00003</b>		INTERNATIONAL FILING DATE <b>JANUARY 4, 2000</b>		PRIORITY DATE CLAIMED <b>JANUARY 5, 1999</b>	
TITLE OF INVENTION <b>METHOD AND DEVICE FOR SIMULTANEOUSLY DETERMINING THE ADHESION, FRICTION, AND OTHER MATERIAL PROPERTIES OF A SAMPLE SURFACE</b>					
APPLICANT(S) FOR DO/EO/US <b>Stifter, Thomas et al.</b>					
Applicant herewith submits to the United States Designated/Elected Office (DO/EO/US) the following items and other information:					
<ol style="list-style-type: none"> <li>1. <input checked="" type="checkbox"/> This is a <b>FIRST</b> submission of items concerning a filing under 35 U.S.C. 371.</li> <li>2. <input type="checkbox"/> This is a <b>SECOND</b> or <b>SUBSEQUENT</b> submission of items concerning a filing under 35 U.S.C. 371.</li> <li>3. <input checked="" type="checkbox"/> This is an express request to begin national examination procedures (35 U.S.C. 371(f)) at any time rather than delay examination until the expiration of the applicable time limit set in 35 U.S.C. 371(b) and PCT Articles 22 and 39(1).</li> <li>4. <input checked="" type="checkbox"/> A proper Demand for International Preliminary Examination was made by the 19th month from the earliest claimed priority date.</li> <li>5. <input checked="" type="checkbox"/> A copy of the International Application as filed (35 U.S.C. 371 (c) (2)) <ol style="list-style-type: none"> <li>a. <input type="checkbox"/> is transmitted herewith (required only if not transmitted by the International Bureau).</li> <li>b. <input checked="" type="checkbox"/> has been transmitted by the International Bureau.</li> <li>c. <input type="checkbox"/> is not required, as the application was filed in the United States Receiving Office (RO/US).</li> </ol> </li> <li>6. <input type="checkbox"/> A translation of the International Application into English (35 U.S.C. 371(c)(2)).</li> <li>7. <input type="checkbox"/> A copy of the International Search Report (PCT/ISA/210).</li> <li>8. <input type="checkbox"/> Amendments to the claims of the International Application under PCT Article 19 (35 U.S.C. 371 (c)(3)) <ol style="list-style-type: none"> <li>a. <input type="checkbox"/> are transmitted herewith (required only if not transmitted by the International Bureau).</li> <li>b. <input type="checkbox"/> have been transmitted by the International Bureau.</li> <li>c. <input type="checkbox"/> have not been made; however, the time limit for making such amendments has NOT expired.</li> <li>d. <input type="checkbox"/> have not been made and will not be made.</li> </ol> </li> <li>9. <input type="checkbox"/> A translation of the amendments to the claims under PCT Article 19 (35 U.S.C. 371(c)(3)).</li> <li>10. <input type="checkbox"/> An oath or declaration of the inventor(s) (35 U.S.C. 371 (c)(4)).</li> <li>11. <input checked="" type="checkbox"/> A copy of the International Preliminary Examination Report (PCT/IPEA/409).</li> <li>12. <input type="checkbox"/> A translation of the annexes to the International Preliminary Examination Report under PCT Article 36 (35 U.S.C. 371 (c)(5)).</li> </ol> <p><b>Items 13 to 20 below concern document(s) or information included:</b></p> <ol style="list-style-type: none"> <li>13. <input type="checkbox"/> An Information Disclosure Statement under 37 CFR 1.97 and 1.98.</li> <li>14. <input type="checkbox"/> An assignment document for recording. A separate cover sheet in compliance with 37 CFR 3.28 and 3.31 is included.</li> <li>15. <input type="checkbox"/> A <b>FIRST</b> preliminary amendment.</li> <li>16. <input type="checkbox"/> A <b>SECOND</b> or <b>SUBSEQUENT</b> preliminary amendment.</li> <li>17. <input type="checkbox"/> A substitute specification.</li> <li>18. <input type="checkbox"/> A change of power of attorney and/or address letter.</li> <li>19. <input checked="" type="checkbox"/> Certificate of Mailing by Express Mail</li> <li>20. <input checked="" type="checkbox"/> Other items or information:</li> </ol>					
<p>Copy of PCT Publication No. WO 00/40946 and filed application text and drawings  PCT/IPEA/416 - Notification of Transmittal of IPER (German)  PCT/IPEA/409 - International Preliminary Examination Report (German)  Application text pages altered during examination process (German)</p>					

U.S. APPLICATION NO. (IF KNOWN, 37 CFR) <b>09/869789</b>		INTERNATIONAL APPLICATION NO. <b>PCT/DE00/00003</b>		ATTORNEY'S DOCKET NUMBER <b>TIL0003</b>	
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21. The following fees are submitted: <b>BASIC NATIONAL FEE ( 37 CFR 1.492 (a) (1) - (5) ) :</b>				<b>CALCULATIONS PTO USE ONLY</b>	
<input type="checkbox"/> Neither international preliminary examination fee (37 CFR 1.482) nor international search fee (37 CFR 1.445(a)(2)) paid to USPTO and International Search Report not prepared by the EPO or JPO ..... <b>\$1,000.00</b>					
<input checked="" type="checkbox"/> International preliminary examination fee (37 CFR 1.482) not paid to USPTO but International Search Report prepared by the EPO or JPO ..... <b>\$860.00</b>					
<input type="checkbox"/> International preliminary examination fee (37 CFR 1.482) not paid to USPTO but international search fee (37 CFR 1.445(a)(2)) paid to USPTO ..... <b>\$710.00</b>					
<input type="checkbox"/> International preliminary examination fee paid to USPTO (37 CFR 1.482) but all claims did not satisfy provisions of PCT Article 33(1)-(4) ..... <b>\$690.00</b>					
<input type="checkbox"/> International preliminary examination fee paid to USPTO (37 CFR 1.482) and all claims satisfied provisions of PCT Article 33(1)-(4) ..... <b>\$100.00</b>					
<b>ENTER APPROPRIATE BASIC FEE AMOUNT =</b>				<b>\$860.00</b>	
Surcharge of <b>\$130.00</b> for furnishing the oath or declaration later than months from the earliest claimed priority date (37 CFR 1.492 (e)). <input type="checkbox"/> 20 <input type="checkbox"/> 30				<b>\$0.00</b>	
CLAIMS	NUMBER FILED	NUMBER EXTRA	RATE		
Total claims	28 - 20 =	8	x \$18.00	<b>\$144.00</b>	
Independent claims	2 - 3 =	0	x \$80.00	<b>\$0.00</b>	
Multiple Dependent Claims (check if applicable). <input checked="" type="checkbox"/>				<b>\$270.00</b>	
<b>TOTAL OF ABOVE CALCULATIONS =</b>				<b>\$1,274.00</b>	
Reduction of 1/2 for filing by small entity, if applicable. Verified Small Entity Statement must also be filed (Note 37 CFR 1.9, 1.27, 1.28) (check if applicable). <input type="checkbox"/>				<b>\$0.00</b>	
<b>SUBTOTAL =</b>				<b>\$1,274.00</b>	
Processing fee of <b>\$130.00</b> for furnishing the English translation later than months from the earliest claimed priority date (37 CFR 1.492 (f)). <input type="checkbox"/> 20 <input type="checkbox"/> 30				<b>\$0.00</b>	
<b>TOTAL NATIONAL FEE =</b>				<b>\$1,274.00</b>	
Fee for recording the enclosed assignment (37 CFR 1.21(h)). The assignment must be accompanied by an appropriate cover sheet (37 CFR 3.28, 3.31) (check if applicable). <input type="checkbox"/>				<b>\$0.00</b>	
<b>TOTAL FEES ENCLOSED =</b>				<b>\$1,274.00</b>	
				Amount to be: refunded	\$
				charged	\$

☒ A check in the amount of **\$1,274.00** to cover the above fees is enclosed.  
  
☐ Please charge my Deposit Account No. \_\_\_\_\_ in the amount of \_\_\_\_\_ to cover the above fees.  
 A duplicate copy of this sheet is enclosed.  
  
☒ The Commissioner is hereby authorized to charge any fees which may be required, or credit any overpayment to Deposit Account No. **02-0385** A duplicate copy of this sheet is enclosed.

**NOTE: Where an appropriate time limit under 37 CFR 1.494 or 1.495 has not been met, a petition to revive (37 CFR 1.137(a) or (b)) must be filed and granted to restore the application to pending status.**

SEND ALL CORRESPONDENCE TO:

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**26,280**  
 REGISTRATION NUMBER

**July 3, 2001**  
 DATE



09869789 PCT/PTO 12 FEB 2002

# 7/a

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re Application of )  
Hans-Ulrich Krottil et al. )  
Serial No.: 09/869,789 ) Group:  
Filed: )  
Title: METHOD AND DEVICE FOR SIMULTANEOUSLY )  
DETERMINING THE ADHESION, FRICTION, AND )  
OTHER MATERIAL PROPERTIES OF A SAMPLE )

**PRELIMINARY AMENDMENT DELETING  
MULTIPLE DEPENDENT CLAIMS**

Assistant Commissioner of Patents  
Washington, DC 20231

Sir:

Please enter the following amendments to the application.

**IN THE CLAIMS**

In claim 3, line 1, delete "or 2".

In claim 5, line 1, delete "or 4".

In claim 6, line 1, delete "or 5".

In claim 7, line 1, delete "or 6".

In claim 9, line 1, delete "one of claims 3 to 8" and substitute therefor --claim 3--.

In claim 11, line 1, delete "one of the preceding claims" and substitute therefor  
--claim 3--.

In claim 13, line 1, delete "one of the preceding claims" and substitute therefor  
--claim 3--.

In claim 17, line 1, delete "one of claims 14 to 16" and substitute therefor --claim 14--.

In claim 19, line 1, delete "or 18".

In claim 21, line 1, delete "one of claims 14 to 20" and substitute therefor --claim 14--.

In claim 23, line 1, delete "one of claims 14 to 22" and substitute therefor --claim 14--.

In claim 24, line 1, delete "one of claims 14 to 24" and substitute therefor --claim 14--.

In claim 25, line 1, delete "one of claims 14 to 24" and substitute therefor --claim 14--.



TRANSLATION

DE 000000003

"Rastersondenmikroskop" / March 21, 2001

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METHOD AND DEVICE FOR SIMULTANEOUSLY DETERMINING THE  
ADHESION, THE FRICTION AND OTHER MATERIAL PROPERTIES OF A  
SAMPLE SURFACE

Specification

[0001] The present invention relates to a method for simultaneously determining at least two material properties, comprising the surface topography, the adhesion, the static and dynamic friction as well as the elasticity and rigidity, by means of a raster probe microscope comprising a raster probe. The invention relates also to an improved raster probe microscope for the carrying-out of the process according to the invention.

[0002] Raster probe microscopy makes possible the non-destructive characterization of sample surfaces on a molecular or atomic scale. Besides the topography of a surface to be examined there can also be determined a number of other surface properties, such as for example the friction, the adhesion, the yieldingness and other elastic properties.

[0003] To the class of the raster probe microscope there belong, for example, the raster tunneling microscope (STM: Scanning Tunneling Microscope), near-field microscope (SNOM: Scanning Near-Field Optical Microscope) and force or raster force microscope (SFM: Scanning Force Microscope) or RKM: Raster Force Microscope).

[0004] Regarding further information on raster probe microscopy let reference be made here to the following publication of Binnig et al, Binnig, G., Quate, C.F. and Gerber, C.: Atomic Force Microscope, Phys. Rev. Lett. 930-933, 56/56 (1986).

[0005] The determination of adhesive forces occurs

[0006] Alternatively to this, adhesive forces can also be determined by a measurement in the so-called pulsed force mode (PFM). With this measuring method the surface of a sample to be examined is scanned periodically in a type of contact mode, with frequencies in the kilohertz range, preferably 0.1 to 3 kHz. By this manner of procedure there can be simultaneously determined also certain other properties of the sample, such as, for example, the local elastic rigidity and the adhesion. The adhesion values are measured on-line. The measuring of the adhesion by means of the pulsed force mode technique has the disadvantage, however, that the measuring speed must be adapted to the vertical modulation frequency and is thus subject to a restriction. With a modulation frequency of 1 kHz and an image resolution of 256 pixels there is yielded, for example, a minimal measuring speed per line of 0.256 s, in order to obtain a new measurement value for each image pixel by a contact between the raster probe and the sample.

[0007] In regard to more detailed information on the



sensitive samples is possible. A carrying-along of sample contaminations can lead to false friction contrasts. Furthermore, sticky sample systems are likewise not measurable.

[0009] Further information on conventional friction force microscopy can be gained, for example, from the following publications: Mate, C.M.; McClelland, G.M.; Erlandson R.; Chiang S.: Atomic-Scale Friction of a Tungsten tip on a Graphite surface, Phys. Rev. Lett. 59, (1987), 1942; Marti, O.; Colcerho, J.; Mylnek, J.; Combined scanning force and friction microscopy of mica, Nanotechnology 1, (1990), 141-144; Meyer, G., Amer, N.M. Simultaneous measurement of lateral and normal or perpendicular forces with an optical-beam-deflection atomic force microscope, Appl. Phys. Lett., (1990), 2098.

[0010] In a relatively new process for the determination of friction by means of a raster probe microscope, the sample to be examined--in addition to a conventional friction measurement in the contact mode (SFFM)--is periodically modulated laterally by means of a shearing piezo element in the 10 kHz range, perpendicularly to the slow scanning direction, in which process the torsion (Tordierung) of the spring beam in the contact mode is recorded by means of the lock-in technique, and from the measurements present there is determined the adhesion and slide friction. Besides, the topography there can be determined also still further mechanical properties, such as the elastic behavior, the (shearing) rigidity and certain relaxation times. For the exact determination of the adhesion and slide friction in dependence on the normal or perpendicular force, however, a measuring series with varying normal forces is necessary. Moreover, a possible alteration or damaging of soft shearing-force-sensitive samples is possible. Furthermore, a carrying along of sample contaminations can lead to wrong friction contacts.

Furthermore, with this, too, sticky sample systems are not measurable at all or are measurable only with great difficulty.

[0011] Further information on this dynamic friction force microscopy can be gained, for example, from the following two literature references: Yamanaka, K. and Tomita, E: Lateral force modulation atomic force microscope for selective imaging of friction forces, Japanese Journal of Applied Physics, Part 1 (Regular Papers & Short Notes), vol. 34, No. 5B, pages 2879-2882, (May 1995); Yamanaka, K., Takano, H.; Tomita, E. and Fujihira, M.: Lateral force modulation atomic force microscopy of Langmuir-Blodgett film in water, Japanese Journal of Applied Physics, Part 1 (Regular Papers, Short Notes & Review Papers), vol. 35, No. 10, pages 5421-5425 (October 1996).

[0012] In the interest of completeness let there also be made reference to the post-published EP 0 896 201 A1, which discloses a raster probe microscope which comprises in addition to a raster probe also a separate detector probe for the detection of the raster probe deflection and for the control of the raster probe distance with respect to a sample surface to be examined. Here the distance dependence of the resonance behavior of the detector probe is used in a vibration excitation of the detector probe and/or of the raster probe by means of an allocated piezoelectric oscillation arrangement.

[0013] A raster probe microscope according to the generic term of claim 1 is known from the publication "Design and calibration of a scanning force microscope for friction, adhesion, and contact potential studies" by Kolestke et al. (Rev. Sci. Instrum., American Institute of Physics, New York (01-09-1995), 66(9), 4566-4574).

[0014] A locally resolved simultaneous measurement of the adhesion forces and of the friction forces and (of further mechanical sample properties) on the surface of a

sample to be examined, is not made possible, however, by any of the measuring processes known from the state of the art. With a single conventional raster force microscopy measurement there can be determined either the adhesion or the friction of the sample to be examined. The determination of both these magnitudes by a single measurement with a raster force microscope has hitherto not yet been technically possible.

[0015] The problem of the present invention lies, therefore, in the creation of an improved raster probe measuring process, with which at least the adhesion and the friction can be measured simultaneously. The magnitudes mentioned should also be measurable here according to possibility, be it alone or in common, with still other properties of interest, such as, for example, certain elastic constants, comprising the adhesion and the rigidity, and/or the topography, in which context the expression "material properties" in the scope of the present specification can cover also optical signals of a sample to be examined, as well as magnetic or electric forces, information data about the temperature distribution and possibly also other measurement values. The problem consists, further, in the creation of a suitable raster probe microscope for the execution of such a measuring process.

[0016] This problem is solved according to the invention by a process in which the raster probe of a raster-probe microscope and/or the sample with the sample surface to be examined is/are moved in vertical and/or horizontal direction so that the raster probe at a pre-determined point of the sample surface interacts in a given manner with the sample surface. The raster probe here is brought into contact preferably with a given perpendicular force with the sample surface. The raster probe and/or the sample is/are subjected to a vertical oscillation and there

is recorded a vertical and/or lateral deformation of a first measuring signal characterizing the vertical and/or the lateral deformation of the raster probe. In addition, a second measuring signal is recorded characterizing the deformation of the raster probe, in which operation the raster probe and/or the sample is/are subjected to a horizontal and/or to a vertical oscillation. The two measuring signals are thereupon evaluated for the determination of the desired sample properties. The first measuring signal serves here for determining the adhesion, while from the second measuring signal, in the manner still to be described in the following, the friction is determined. In order to obtain a complete information about the area of the sample surface to be examined, the raster probe and/or the sample is/are moved again, in order in the above-described manner to bring the raster probe into contact with the sample surface at the next point to be examined, at which the aforescribed measuring process is repeated. In this manner the entire surface area to be examined is scanned linearly, as is familiar to specialists in this field.

[0017] Hereby there is obtained not only a topographic image of the sample, but it is possible for the first time, with one and the same measurement, to locally determine the friction, the adhesion and certain elastic properties of a sample on a molecular level down to the atomic level. The friction can be determined here simultaneously, in dependence on diverse normal or perpendicular forces so that measuring series with varying normal or perpendicular forces are reduced to a single measurement. This is associated not only with a clearly lower burdening of the sample by a scanning (Abrastern) but by reason of the temporally same environmental conditions (temperature, air humidity, etc., sample aging), it makes possible also a better comparability of the determined friction values. By a varying of the



phase displacement). Hereby statements about the mechanical behavior of the sample are possible, especially about its elasticity and rigidity.

**[0019]** Advantageously the raster probe and/or the sample is excited in vertical direction with a frequency of at least 10 Hz and an amplitude of at least 1 nm, the preferred frequency and amplitude range being 500 Hz to 1 kHz and 10 to 500 nm, respectively.

**[0020]** To the vertical oscillation of the sample and/or the probe there is preferably superimposed at least one second oscillation with a frequency of at least 1 kHz and an amplitude of at least 0.1 nm, in particular, however, with a frequency of 5 kHz to 1 MHz and an amplitude of 1 to 10 nm. There is advantageously used a horizontal oscillation with a frequency of at least 500 Hz, in particular, however, 10 to 100 kHz, and an amplitude of at least 0.1 nm, especially, however, 1 to 30 nm.

**[0021]** In a preferred example of execution the second measuring signal is evaluated in the manner thoroughly to be described below by means of a lock-in amplifier by which over a Fourier transformation of the measuring signal there are determined the friction amplitude and the phase. As raster probe there is used in particular the point or tip of a force microscope and/or of an optical near-field microscope with which there can also be detected optical signals of a sample to be examined, in which context the point or tip of the force microscope and the point or tip of the optical near-field microscope can also be integrated in a common raster probe.

**[0022]** A raster probe microscope suited for the execution of this process according to the invention, with the features given in the generic term of claim 1, additionally also comprises according to the invention an arrangement for the vertical and/or horizontal moving of the sample, an arrangement for detecting the sample movement and

an arrangement for detecting the vertical and/or lateral deformation of the raster probe. The arrangements for the moving of the raster probe or of the sample are formed here in such manner that the raster probe and the sample surface are bringable into contact or are brought into contact in such manner that they interact with one another in a determined manner, which comprises in particular a contact with a given normal or perpendicular force.

**[0023]** In a preferred form of execution the arrangements for moving the raster probe or the sample comprise at least one piezo element, at least one piezo element being provided preferably for each of these arrangements.

**[0024]** The arrangements for moving the raster probe or the sample, especially the piezo elements mentioned, are preferably periodically excited or modulated, it being already possible to learn the type of excitation or modulation from the above description of the inventive process.

**[0025]** The arrangement for detecting the vertical and/or lateral deformation of the raster probe may comprise, in a preferred constructive execution, a mirror applied for example by a corresponding coating to the raster probe, which mirror is designed for the deflection of an incident laser beam, the resulting deflection being able to serve as measure for the deformation of the raster probe that is present. Corresponding information can be gained, however, for example also capacitively, interferometrically or piezoelectrically.

**[0026]** The inventive raster probe microscope preferably comprises an evaluating arrangement for the simultaneous determination of at least two material properties, comprising the adhesion, the static and dynamic friction, the surface topography as well as the elasticity and rigidity, by evaluation of the detected deformation of



- 4;
- Fig. 7A-7H the principle of a combination pulsed-force-mode/force modulation;
- Fig. 8 a run-off diagram for a measurement according to the invention for the combination pulsed-force-mode/force modulation according to Fig. 7;
- Fig. 9A-9B theoretically calculated friction amplitudes and phase-dependencies of the cantilever on the modulation amplitudes  $A_M$  of an excitation shearing piezo element;
- Fig. 10 measuring signals of the new-type raster probe microscope on the basis of a vertical bending of the cantilever (combination pulsed-force-mode/force modulation);
- Fig. 11 measuring signals of the new-type raster probe microscope on the basis of a vertical bending of the cantilever (combination pulsed-force-mode/force modulation);
- Fig. 12A-12D a depiction of the topography of the adhesion and of the friction on a sample surface, examined with a raster probe microscope according to the invention for a combination pulsed-force-mode/dynamic friction; and
- Fig. 13A-13D a depiction of the topography, the adhesion, the friction on a sample surface, examined with a raster probe microscope according to the invention for a combination pulsed-force-mode/force modulation.

[0029] The raster probe microscope represented in Fig. 1 comprises a raster probe 1 with a beam or cantilever 3 and a measuring point or tip 5. The measuring point 5 can be made, for example, of silicon or silicone nitride, for example  $\text{Si}_3\text{N}_4$ . The vertical shifting of the raster probe 1 is performed with the aid of a piezo element 7. The

measurement of the deformation of the beam 3, which in the vertical process is a measure for the adhesion forces, is determined by means of a laser structure (not shown in detail). For this the light 9 of an unrepresented laser light source is projected onto the beam 3, from which it is reflected so that the reflected beam 11 impinges on a measuring arrangement 13, which can comprise, for example, the photosensitive layer of a segmented photo-diode.

[0030] According to the position of the beam 3 the light beam 11 is deflected upward or downward and to the left, or to the right, respectively, from the zero position shown in the drawing, and it is transformed by the photosensitive layer of the measuring arrangement 13 into an electrical signal. The electrical measuring signal yielded in a relative movement occurring in horizontal direction between the beam 3 or the measuring point or tip 5 and the sample surface 30--which (signal) according to Fig. 3B generates an essentially laterally occurring deflection of the reflected light beam 11 and is designated in the following as second measuring signal--is transmitted over a line 15 to a lock-in amplifier 17, in which a Fourier transformation is carried out and there the real part and the imaginary part of the signal are determined, from which (in a manner still to be described in the following) the desired friction can be determined. The electrical measuring signal yielded in a vertical relative movement between the measuring point or tip 5 and the sample surface 30 (the so-called first measuring signal, in which the reflected light beam according to Fig. 3A is deflected essentially upward or downward) is delivered for the determination of the adhesion over an unrepresented line, directly to a likewise not represented evaluating arrangement 112 (see Fig. 6).

[0031] Let it be remarked that the detection of the deformation of the beam 3 by a photo-indicating principle of

the type represented is only one possible type of detection, and that for specialists in this field, theoretically other possibilities of detection are also conceivable, such as, for example capacitive, interferometric or piezoelectric detection possibilities.

**[0032]** Besides the vertical movability, on the sample table 23 that carries the sample 25 there is arranged, according to the invention, a piezo element (not shown), with which the sample table 23 and therewith also the sample 25 can also be moved or excited laterally as was already mentioned above, for the determination of the static and dynamic friction. The measuring beam 3 of the force point or tip 5 is twisted and/or bent, as shown in broken lines, in which operation the torsion and/or bending arising is a measure of the friction forces present.

**[0033]** In Fig. 2 the coordinate system is again clarified. There is shown the measuring point or tip 5 which is moved with respect to the surface 30. A raising and lowering of the measuring point or tip 5 and/or of the sample 25 in Z-direction, as indicated with arrow 32, makes possible the adhesion measurement in the point 34; a moving or exciting in the plane of the same surface 30 along the arrow makes possible the measurement of the static and dynamic friction at the point 34 shown in the drawing.

**[0034]** In Figs. 3A and 3B the resulting measuring signals are once again shown in detail. In the vertical movement in Z-direction for the measurement of the adhesion the beam 3 according to Fig. 3A is bent in Z-direction. The light beam coming from a laser 26 is reflected from the beam 3 and deflected essentially upward or downward on the measuring arrangement or measuring surface 13, the resulting deflection being a measure of the adhesion force. In Fig. 3B the measuring signal for a friction measurement is shown, in which the raster probe 1 and the sample 25 are subjected to a relative vertical movement. Again, the light beam 9 of

the laser 38 is led onto the beam 3 and from this it is reflected on the measuring surface 13 essentially to the left or to the right. Since the beam 3 is twisted and/or bent by the friction of the measuring point or tip on the surface 30, the resulting deflection is a measure of the friction force.

**[0035]** Figures 4A to 4H show, with the aid of a representation of the different beam or cantilever deformations during a period for the detection of the local material properties at a certain sample point, the principle of a combination pulsed-force-mode/dynamic friction, in which for better perspicuity it is only in Fig. 4A that reference numbers are given. The diagrams shown in the individual figures show here the time dependence of the detected measuring signals, in which the curves 1 and 2 correspond to the real part  $x$  and to the imaginary part  $y$ , respectively, of the second measuring signal, already mentioned above, on the basis of the lateral cantilever deformation, while the curve 3 shows a typical pulsed-force-mode force signal (first measuring signal).

**[0036]** In Fig. 4A the raster probe 1 is still so far remote from the sample 25 to be examined, that there is still no interaction between the raster probe 1 and the sample 25. The detected measuring signals, therefore, are equal to zero at this point in time.

**[0037]** In the further approach of the raster probe 1 to the sample 25 shown in Fig 4B, the raster probe 1 or the measuring point or tip 5, by reason of the negative (attractive) force between the raster probe 1 and the sample 25, comes in contact with the sample surface 30. This leads to a "snap-on" peak in the pulsed-force measuring signal (first measuring signal) represented in curve 3.

**[0038]** The raster probe 1 is pressed further onto the sample 25, which according to Fig. 4C leads to a rise of the first measuring signal (curve 30. Simultaneously the raster

probe 1 is horizontally deformed by reason of the horizontal modulation. This leads to the detection of a second measuring signal that is represented broken down into real part and imaginary part (curves 1 and 2, respectively).

**[0039]** Fig. 4D illustrates how the raster probe 1 is pressed onto the sample 25 until a given normal or perpendicular force that is being registered is reached. The positive repulsive force reaches a maximum value, so that both the first measuring signal (curve 3) and also the second measuring signal (curves 1 and 2) take on a maximal value.

**[0040]** In Fig. 4E the raster probe 1 is again withdrawn from the sample 25, so that the detected measuring signals become smaller.

**[0041]** In a further withdrawal of the raster probe 1 from the sample surface 30 the measuring or force signals become still smaller (see Fig. 4F) and one comes again into the attractive range by reason of the adhesive interaction between the measuring point or tip 5 and the sample 25.

**[0042]** Fig. 4G illustrates how the measuring point or tip 5 still remains sticking to the sample 25 and how the negative force needed for the separation of that measuring point or tip from the sample 25, which is here designated as adhesion force, becomes maximal.

**[0043]** In a further withdrawal of the measuring point or tip 5 from the sample surface 30 that measuring point 5 finally comes out of contact with the sample surface 30 and swings out, which makes itself noticeable by a dying-out in the pulsed-force mode measuring signal 3 (see Fig. 4H). A new period begins.

**[0044]** Fig. 5 illustrates the principle of a dynamic friction measurements. In each modulation period a full friction loop is completed. At a certain deflection the measuring point or tip 5 of the force microscope can follow

the excitation modulation; one finds oneself in the adhesion friction range. With a further increase of the modulation amplitude the measuring point or tip 5 of the force microscope can no longer follow the deflection and sliding friction sets in, and the detected amplitude decreases.

**[0045]** Fig. 6 shows a process diagram for the combination of pulsed-force mode and dynamic friction, already represented in Fig. 4, which makes possible a simultaneous measurement of the adhesion and friction or elasticity. First of all the raster probe 1 is moved with the aid of a control arrangement 100 to a predetermined sample point X, Y. Then, with the aid of a function generator 102 a periodic modulation voltage is generated which results in a periodic vertical movement of the raster probe 1 at a frequency of at least 10 Hz, especially, however, 500 Hz to 2 kHz, and an amplitude of at least 1 nm, but in particular 10 to 500 nm. In addition to the vertical periodic movement, with the aid of a function generator 104 the sample 25 is moved periodically laterally, in which process the frequency amounts to at least 500 Hz, especially 10 to 100 kHz, and the amplitude to at least 0.1 nm but in particular 1 to 30 nm. For this there is used a modulation piezo element 106. The detector arrangement 108, already represented above, which detects both the torsion (Tordierung) and/or the bending of the raster probe 1, delivers over a line 109 to a lock-in amplifier 110 the above-described second measuring signal, which is subjected to a Fourier transformation and is split into a real part x and an imaginary part y of the deformation of the raster probe.

**[0046]** In a likewise possible combination pulsed-force mode/force modulation, for which a corresponding run-off diagram is shown in Fig. 8, the raster probe 1 is first moved, likewise with the aid of the control arrangement 100, onto a pre-determined sample point x,y. Then, with the aid



(AAS), while Fig. 9B shows a resulting amplitude-phase spectrum (APS) with the dependence of the detected phase on the modulation amplitude  $A_M$ . For very high modulation- or excitation-amplitudes  $A_M$ , there holds

$$\lim_{A_M \rightarrow \infty} r = 4 \cdot F_G / \Pi$$

In the phase spectrum APS there is to be expected in the sliding friction range, according to q-factor ( $q = F_G / F_H$ ) a clear phase displacement, in which  $F_G$  is the sliding friction and  $F_H$  the adhesion friction.

**[0050]** The friction coefficients  $\mu$  are determined over  $\mu = F_G / F_N$ , in which the normal or perpendicular force  $F_N$  is obtained from the calibrated pulsed-force-mode measuring signal on which the regulation is performed. An analogous statement holds for the APS.

**[0051]** If dynamic friction measurements are carried out with constant modulation amplitudes  $A_M$ , then to the qualitative friction and phase contrasts by means of AAS and APS, respectively, there can be allocated quantitative friction values and phase displacements.

**[0052]** The recorded first measuring signals are transmitted over the line 111 directly onto the evaluating arrangement 112.

**[0053]** From the recorded force signals from the evaluating arrangement 112, by means of a microcomputer with a suitable computer program, there are determined also the adhesion 114 and the elasticity 116.

**[0054]** If this was carried out for a certain XY-point of the sample, then with the aid of the control arrangement 100 the raster probe 1 is moved to another point X, Y of the sample surface 30. At this point the previously described measurement is repeated. As was already described above, in this manner the complete sample 25 is raster-scanned (abgerastert), in which operation there is obtained, besides

the topography, a complete picture of the adhesion, of the friction and of the elasticity on the sample surface 30.

**[0055]** The vertical and/or horizontal modulation can be generated not only by a shearing piezo element mounted under the sample 25 but, for example, also by a scanning piezo element.

**[0056]** In Fig. 10 there are shown the measuring signals detected at a pre-determined sample point on the basis of the lateral and vertical bending and/or torsion (Tordierung) of the beam or cantilever 3, which were obtained with the process of the invention by a combination pulsed-force mode/dynamic friction. After an electronic processing and the selection of certain characteristic measuring values it is possible from these measuring signals, in the manner described, to determine the desired sample-specific properties. The curve 200 shows the real part  $x$  of a recorded second measuring signal issued from the lock-in amplifier 110, on the basis of the deformation of the cantilever, while the curve 202 represents the imaginary part of this measuring signal. The curve 204 shows the detected pulsed-force measuring signal (the measuring signal 1) on the basis of the vertical bending of the cantilever 3.

**[0057]** Fig. 11 shows a corresponding representation for a combination pulsed-force-mode/force modulation.

**[0058]** In Fig. 12 there are represented images of a sample surface 30, recorded with the inventive process, in which the horizontal excitation frequency of the sample amounts to 93 kHz and the vertical excitation frequency of the probe 1 amounts to 1 kHz. Fig 12A shows the sample topography that is obtained from a force regulating. Fig 12B shows the adhesion to the sample surface 30 and Fig. 12C the friction amplitude on the sample surface 30. In Fig. 12D, finally, there is shown the phase of the measuring signal.



## Patent Claims

1. Raster probe microscope for the examination of sample surfaces with:
  - a raster probe (1),
  - a holding device (23) for a sample (25) with the sample surface (30) to be examined;
  - an arrangement (106) for moving the raster probe (1) and/or the sample (25) by which the raster probe (1) and/or the sample (25) are bringable into contact or are brought into contact so that they interact with one another in a given manner; and with an arrangement for detecting the movement of the raster probe and/or sample; characterized by an arrangement (100, 102, 104) for controlling the movement of the raster probe and/or sample and for exciting a vertical first raster-probe and/or sample oscillation as well as a vertical and/or horizontal second raster-probe and/or sample oscillation; and
  - an arrangement (103, 108) for detecting a vertical and/or a lateral deformation of the raster probe (1) in a vertical first or in a vertical and/or horizontal oscillation excitation and for the recording (Aufnehmen) of two measuring signals characterizing the deformation of the raster probe (1) in a vertical first or a vertical and/or horizontal second oscillation excitation of the raster probe (1) and/or of the sample (25).
2. Raster probe microscope according to claim 1, characterized in that the arrangement for moving the raster probe (1) and/or the sample (25) comprises at least one first piezo element.
3. Raster probe microscope according to claim 1 or 2, characterized by periodic raster-probe and/or sample

oscillations.

4. Raster probe microscope according to claim 3,  
characterized in that the oscillation direction runs parallel or perpendicular to the sensing (Abtast-) or scanning direction.
5. Raster probe microscope according to claim 3 or 4,  
characterized in that the vertical oscillation of the raster probe (1) and/or of the sample (25) occurs with a first frequency of at least 10 Hz and a first amplitude of at least 1 nm.
6. Raster probe microscope according to claim 4 or 5,  
characterized in that the frequency amounts to 500 Hz to 1 kHz and the amplitude to 10 to 500 nm.
7. Raster probe microscope according to claim 5 or 6,  
characterized in that the vertical oscillation of the raster probe (1) and/or of the sample (25) is additionally excited or modulated with a second frequency of at least 1 kHz and a second amplitude of at least 0.1nm.
8. Raster probe microscope according to claim 7,  
characterized in that the frequency ranges from 5 kHz to 1 MHz and the amplitude from 1 to 10 nm.
9. Raster probe microscope according to one of claims 3 to 8,  
characterized in that the sescond raster-probe and/or sample oscillation is a horizontal oscillation with a frequency of at least 500 Hz and an amplitude of at least 0.1 nm.
10. Raster probe microscope according to claim 7,  
characterized in that the frequency ranges from 10 to 100 kH and the amplitude from 1 to 30 nm.
11. Raster probe microscope according to one of the preceding claims, characterized in that an evaluating arrangement (17) for the two measuring signals for the simultaneous determination of at least two material properties, comprising the adhesion, the static and

dynamic friction, the surface topography as well as the elasticity and rigidity.

12. Raster probe microscope according to claim 11, characterized in that the evaluating arrangement comprises a lock-in amplifier (17) and/or a microcomputer (112).
13. Raster probe microscope according to one of the preceding claims, characterized in that the raster probe (1) is a point or tip (5) of a force microscope and/or of an optical near-field microscope.
14. Process for the simultaneous determination of at least two material properties, comprising the adhesion, the static and dynamic friction, the surface topography as well as the elasticity and rigidity of the surface of a sample (25) to be examined, by means of a raster probe microscope comprising a raster probe (1) with the following process steps:
  - 14.1 Moving the raster probe (1) and/or the sample (25) with the sample surface (30) to be examined until the raster probe (1) at a pre-determined point (34) of the sample surface (30) to be examined, interacts in a determined manner with the sample surface (30), wherein the raster probe (1) and/or the sample (25) is subjected to a vertical first oscillation;
  - 14.2 Recording of a first measuring signal characterizing the deformation of the raster probe (1);
  - 14.3 Recording of a second measuring signal characterizing the deformation of the raster probe (1), in which the raster probe (1) and/or the sample (25) is subjected to a horizontal and/or vertical second oscillation;
  - 14.4 Determination of the desired material properties from the two measuring signals; and

- 14.5 Scanning of the region of the sample surface (30) to be examined by a return to the process step 14.1.
15. Process according to claim 14, characterized in that the raster probe (1) and/or the sample (25) is subjected to at least one periodic oscillation.
  16. Process according to claim 15, characterized in that the oscillation direction is chosen perpendicular to or parallel to the sensing or scanning direction.
  17. Process according to one of claims 14 to 16, characterized in that the vertical oscillation or the vertical oscillations has/have a frequency of at least 10 Hz and an amplitude of at least 1 nm.
  18. Process according to claim 17, characterized in that the frequency ranges from 500 Hz to 2 kHz and the amplitude from 10 to 500 nm.
  19. Process according to claim 17 or 18, characterized in that on the vertical oscillation (or oscillations) there is superimposed at least one second oscillation with a frequency of at least 1 kHz and an amplitude of at 0.1 nm.
  20. Process according to claim 19, characterized in that the frequency ranges from 5 kHz to 1 Mhz and the amplitude from 1 to 10 nm.
  21. Process according to one of claims 14 to 20, characterized in that the horizontal oscillation has a frequency of at least 500 Hz and an amplitude of at least 0.1 nm.
  22. Process according to claim 21, characterized in that the frequency ranges from 10 to 100 kHz and the amplitude from 1 to 30 nm.
  23. Process according to one of claims 14 to 22, characterized in that the raster probe (1) is brought into contact with the sample surface (30) with a determined normal or perpendicular force.

24. Process according to one of claims 14 to 24,  
characterized in that for the evaluation of the  
measuring signals there is used a lock-in amplifier  
(17, 110) and/or a microcomputer (112).
25. Process according to one of claims 14 to 24,  
characterized in that as raster probe (1) there is used  
the point or tip (5) of a force microscope and/or of an  
optical near-field microscope.
26. Process according to claim 25, characterized in that  
the point or tip (5) of the force microscope and the  
point or tip of the optical near-field microscope are  
integrated in a common raster probe (1).
27. Process according to one of claims 14 to 26,  
characterized in that the raster probe (1) and/or the  
sample (15) are subjected simultaneously at least to a  
vertical and at least to a horizontal oscillation.

## Abstract

A process is described for the location-resolved simultaneous detection of the adhesion and friction as well as possibly of other material properties of a sample surface (30) to be examined by means of a raster probe microscope comprising a raster probe (1). The raster probe (1) and/or the sample (25) with sample surface (30), are moved here until at a point (34) of the sample surface (30) to be examined the raster probe (1) interacts in a determined manner with this surface. The raster probe (1) and/or the sample (25) are subjected here to a vertical oscillation, and a first measuring signal characterized by the deformation of the raster probe (1) is recorded. Furthermore, a second measuring signal characterizing the deformation of the raster probe (1) is recorded, wherein the raster probe (1) and/or the sample (25) are subjected to a horizontal and/or vertical oscillation. From these two measuring signals, now, by means of a suitable evaluating arrangement the desired material properties are determined. For the detection of the entire surface area to be examined the raster probe (1) and or the sample (25) are again moved and for the repetition of the measuring process described brought into contact with the sample surface (30) in the above described manner. A suitable raster probe microscope is described for the execution of this process.

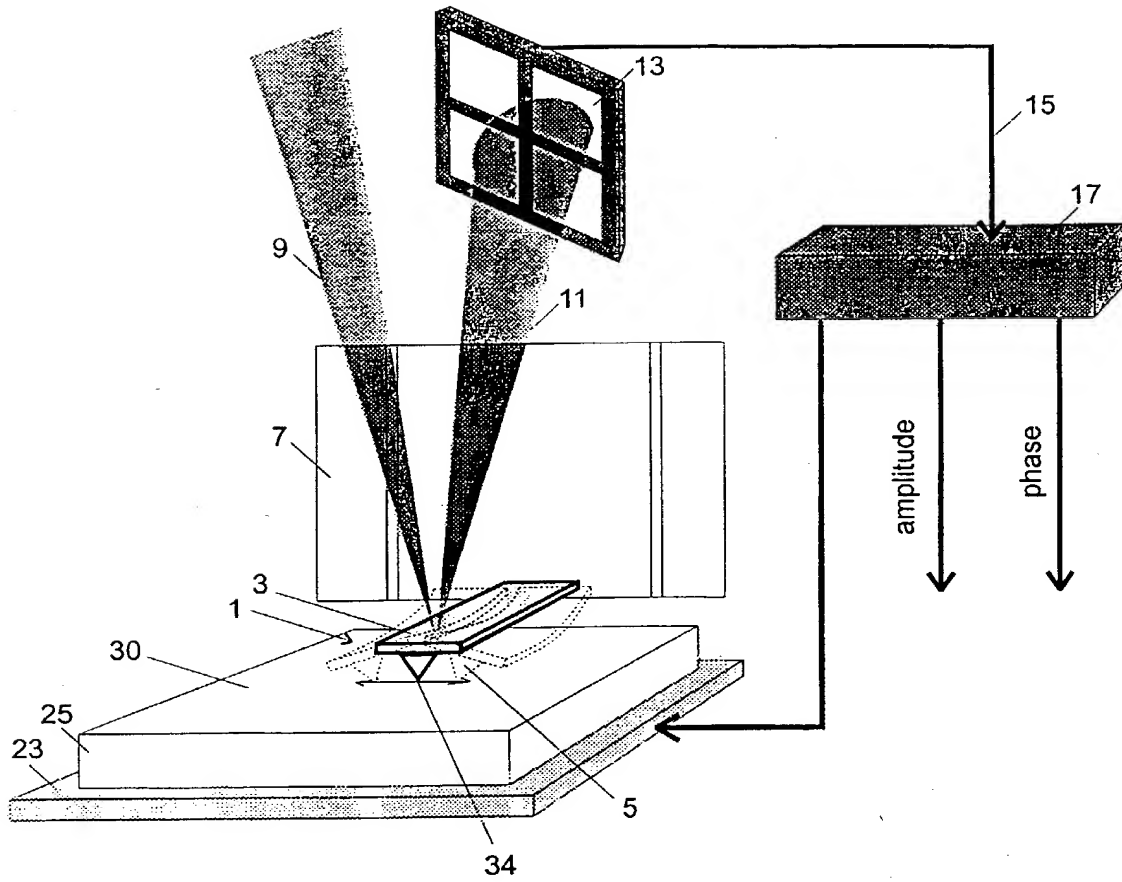


Fig. 1

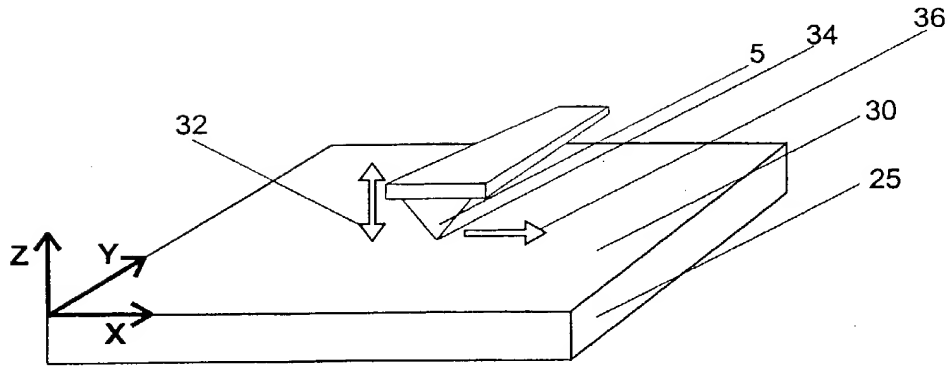


Fig. 2

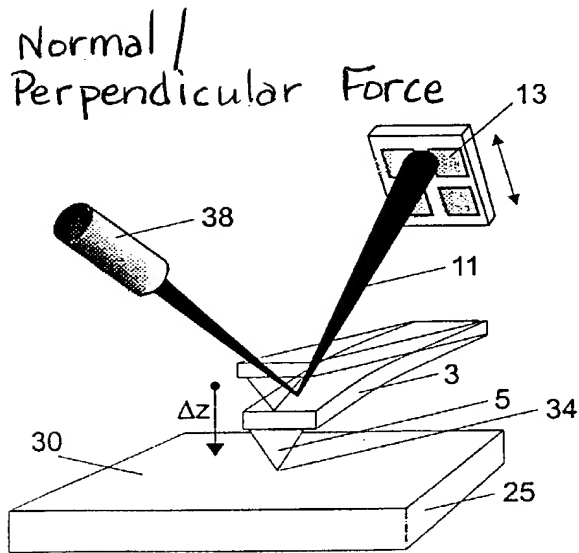


Fig. 3A

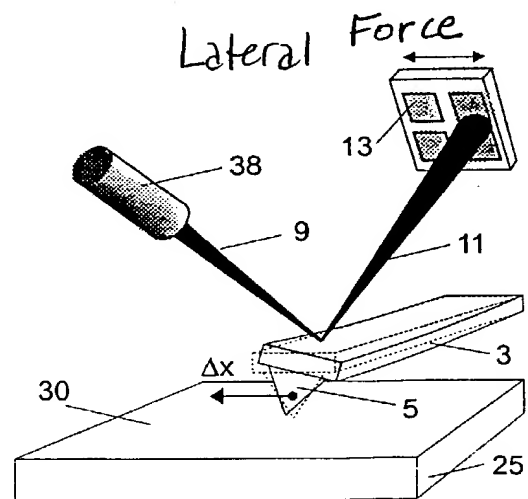
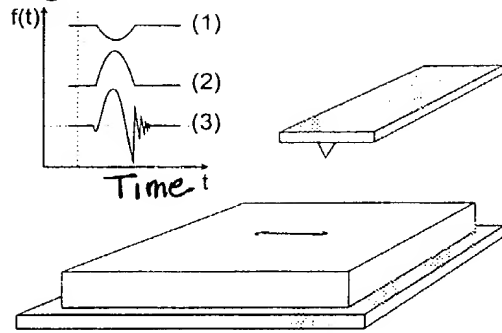
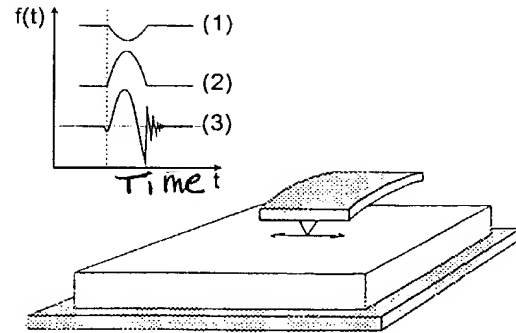


Fig. 3B

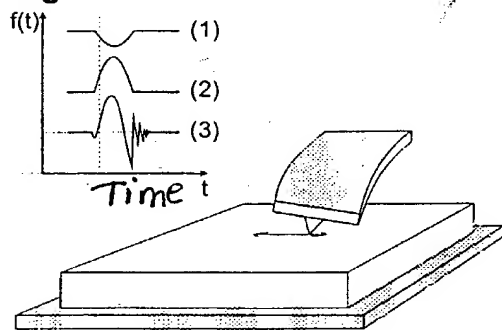
**Fig. 4A**



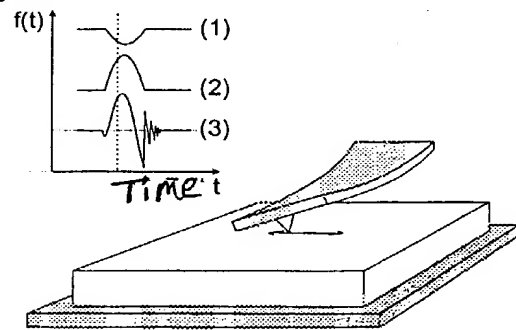
**Fig. 4B**



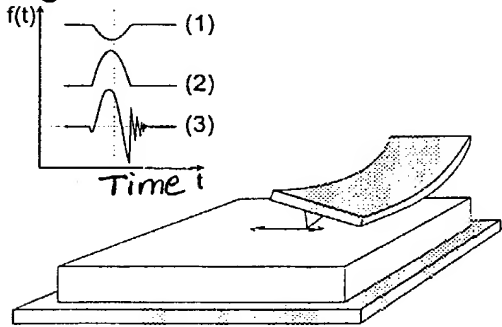
**Fig. 4C**



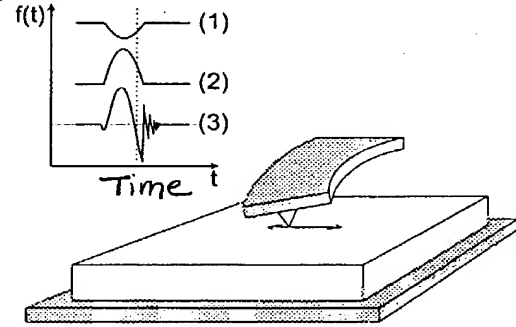
**Fig. 4D**



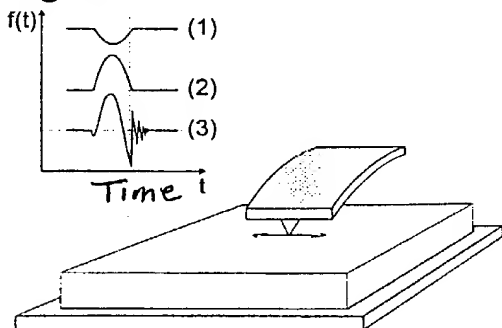
**Fig. 4E**



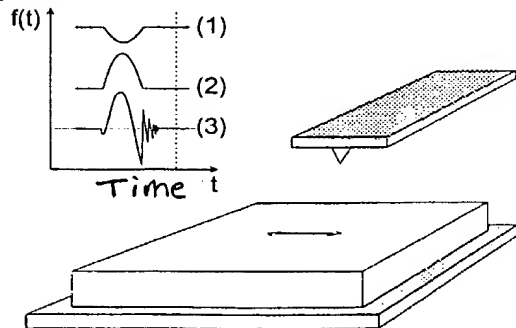
**Fig. 4F**



**Fig. 4G**



**Fig. 4H**



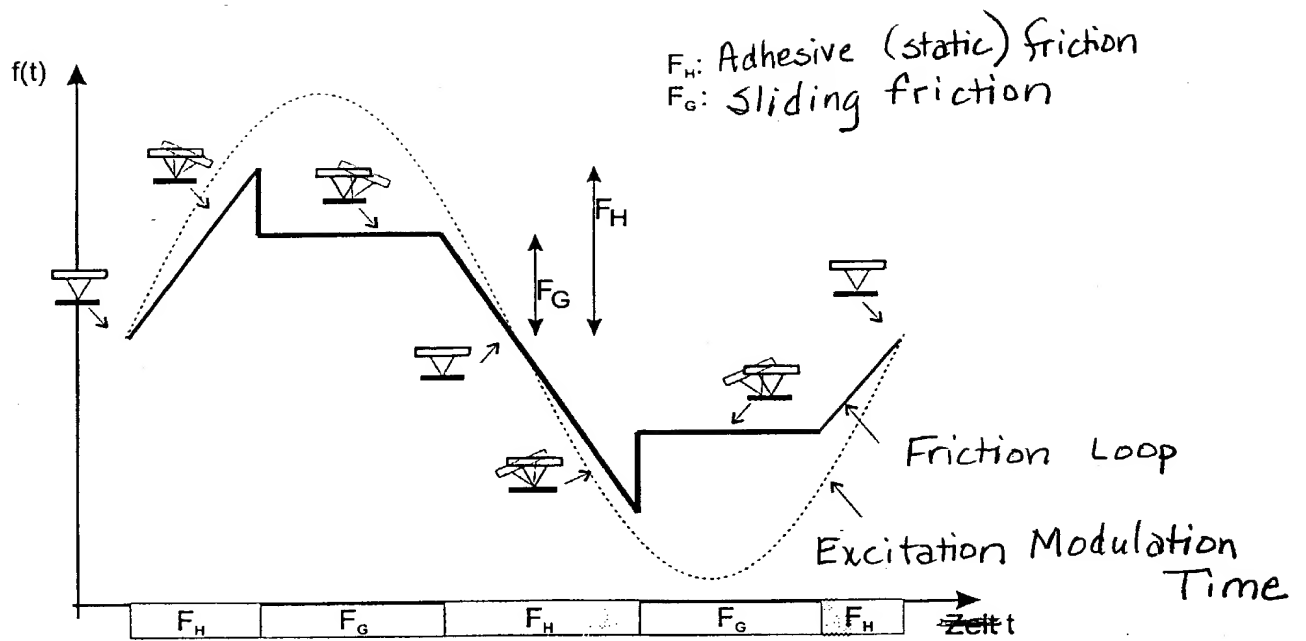


Fig. 5

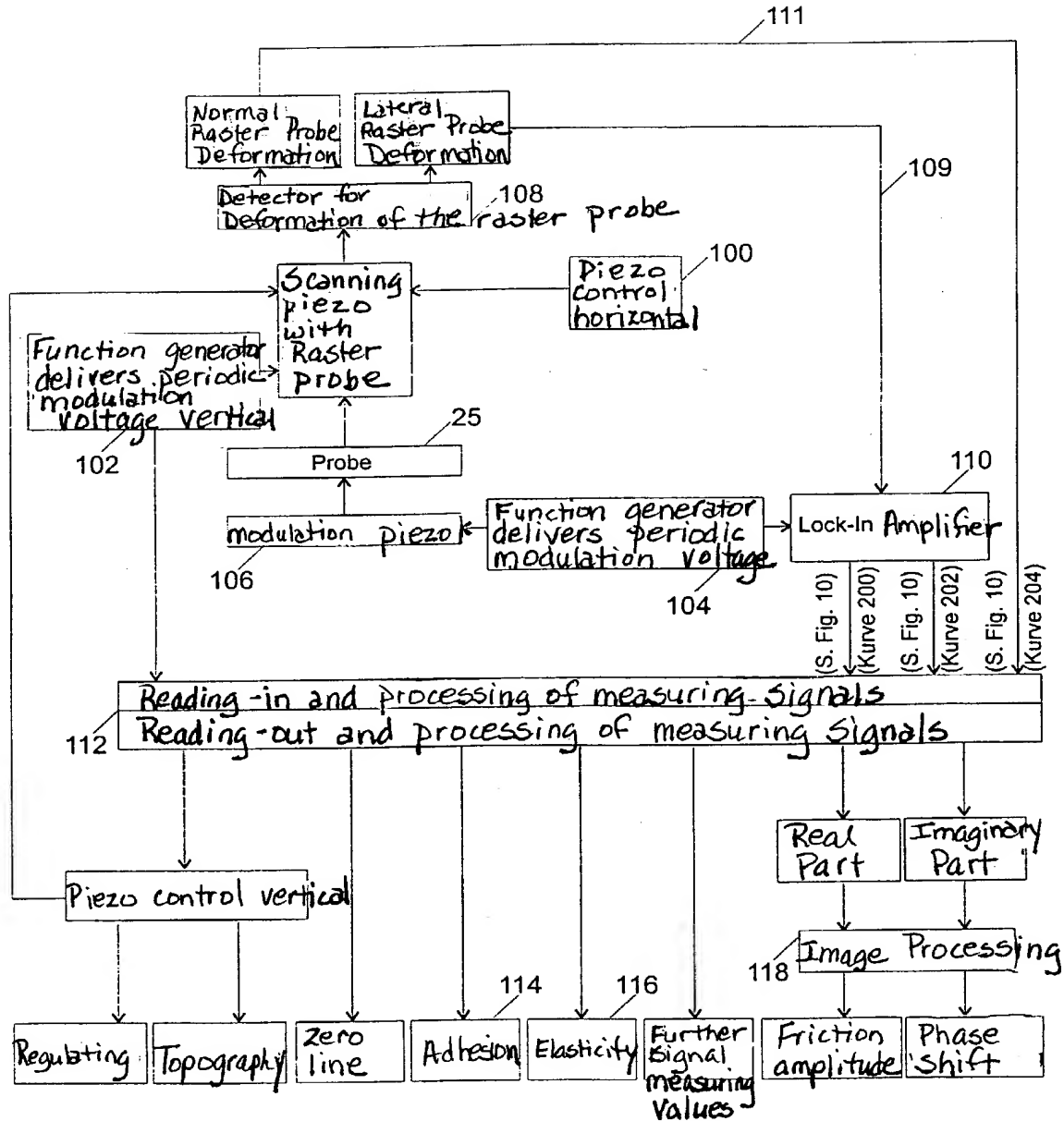


Fig. 6

Fig. 7A

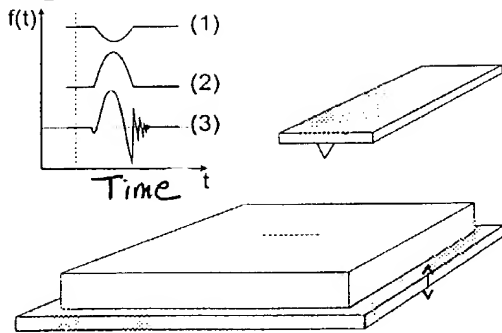


Fig. 7B

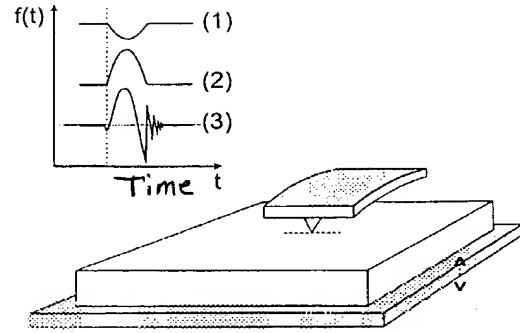


Fig. 7C

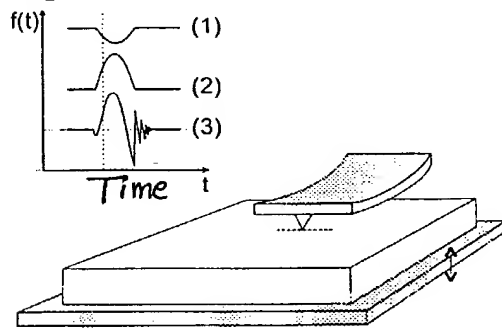


Fig. 7D

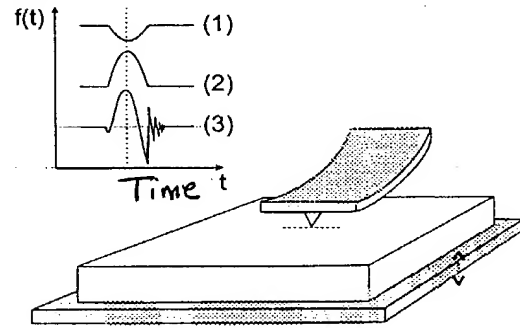


Fig. 7E

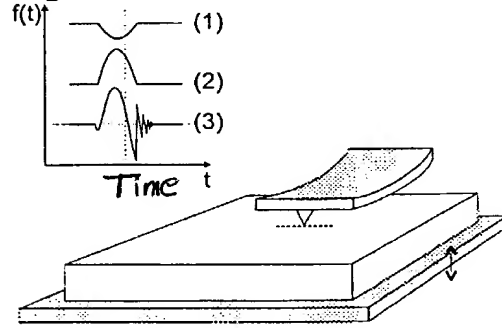


Fig. 7F

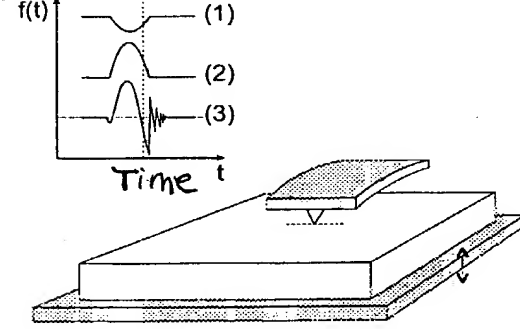


Fig. 7G

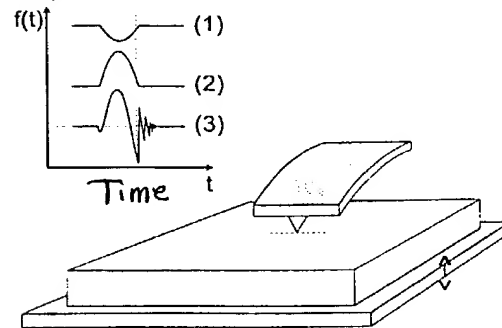
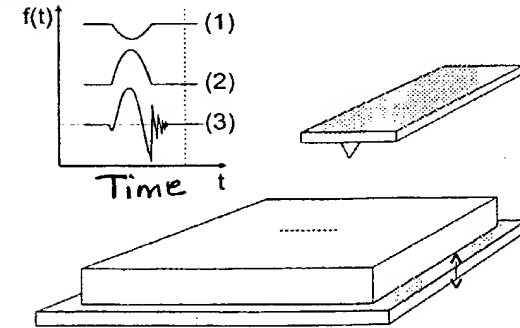


Fig. 7H



**Fig. 8**

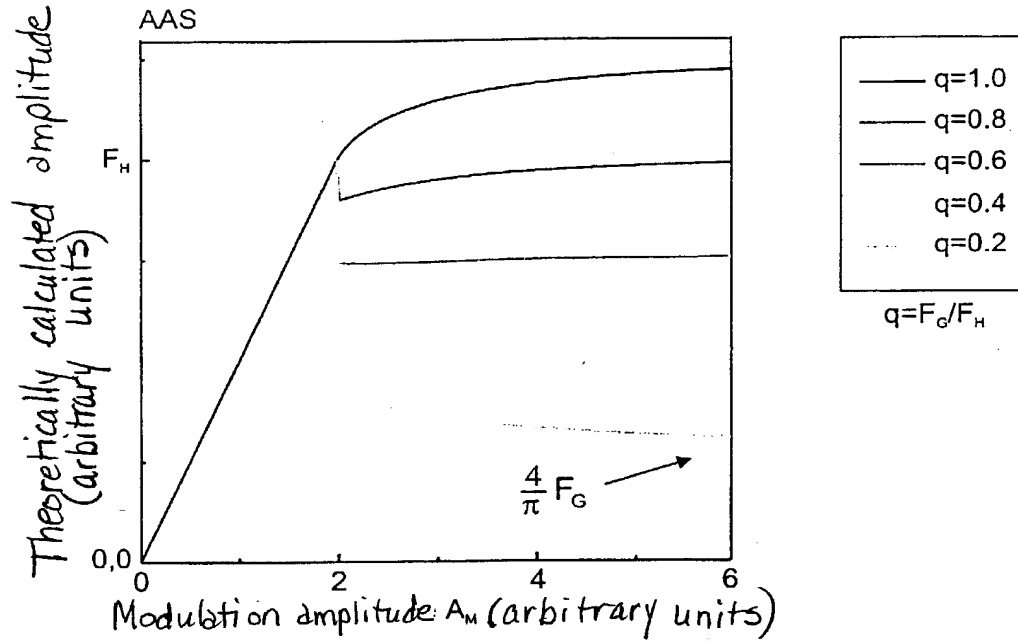


Fig. 9A

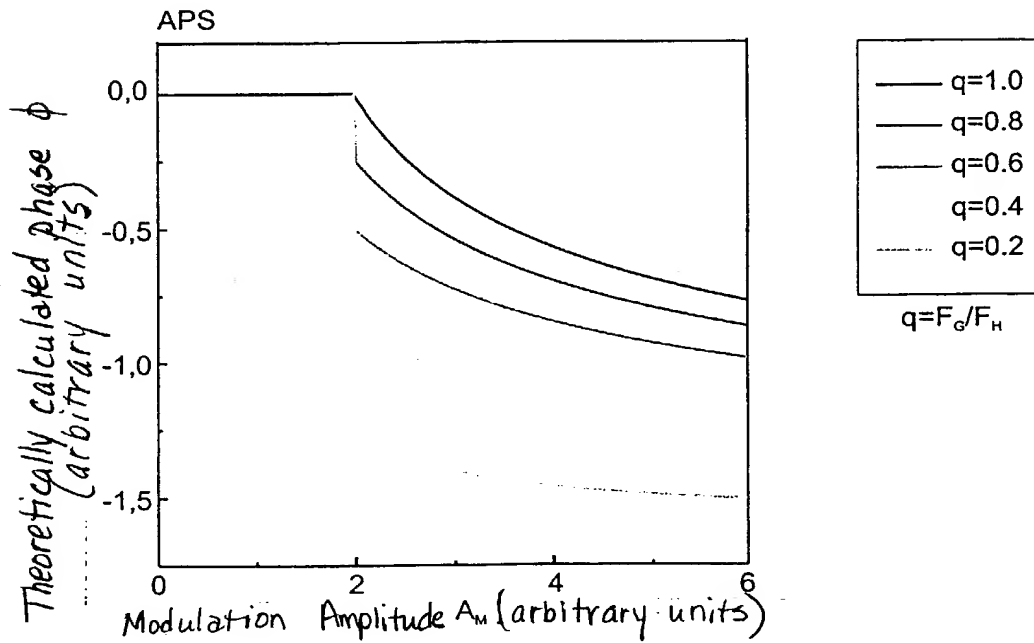


Fig. 9B

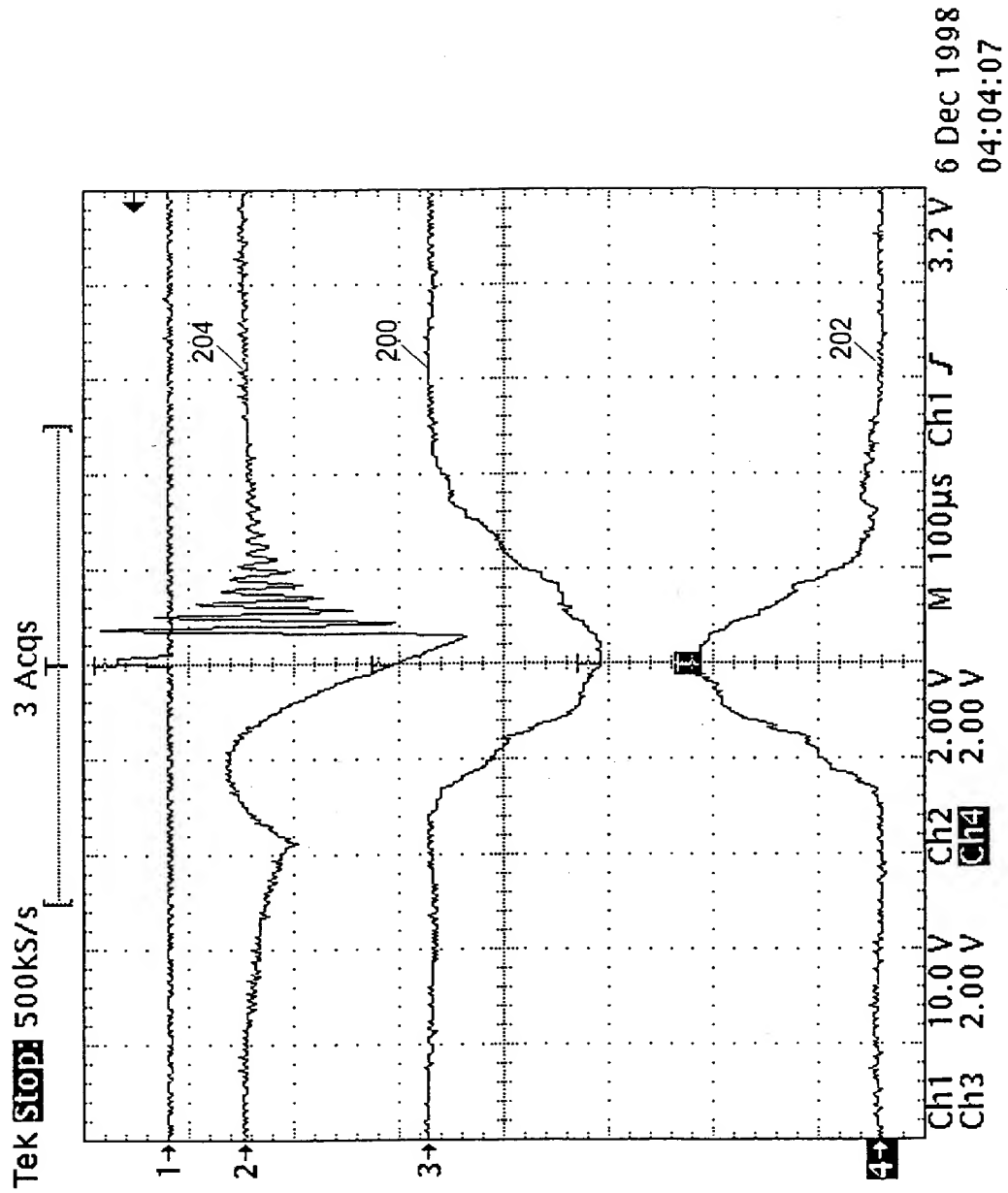


Fig. 10

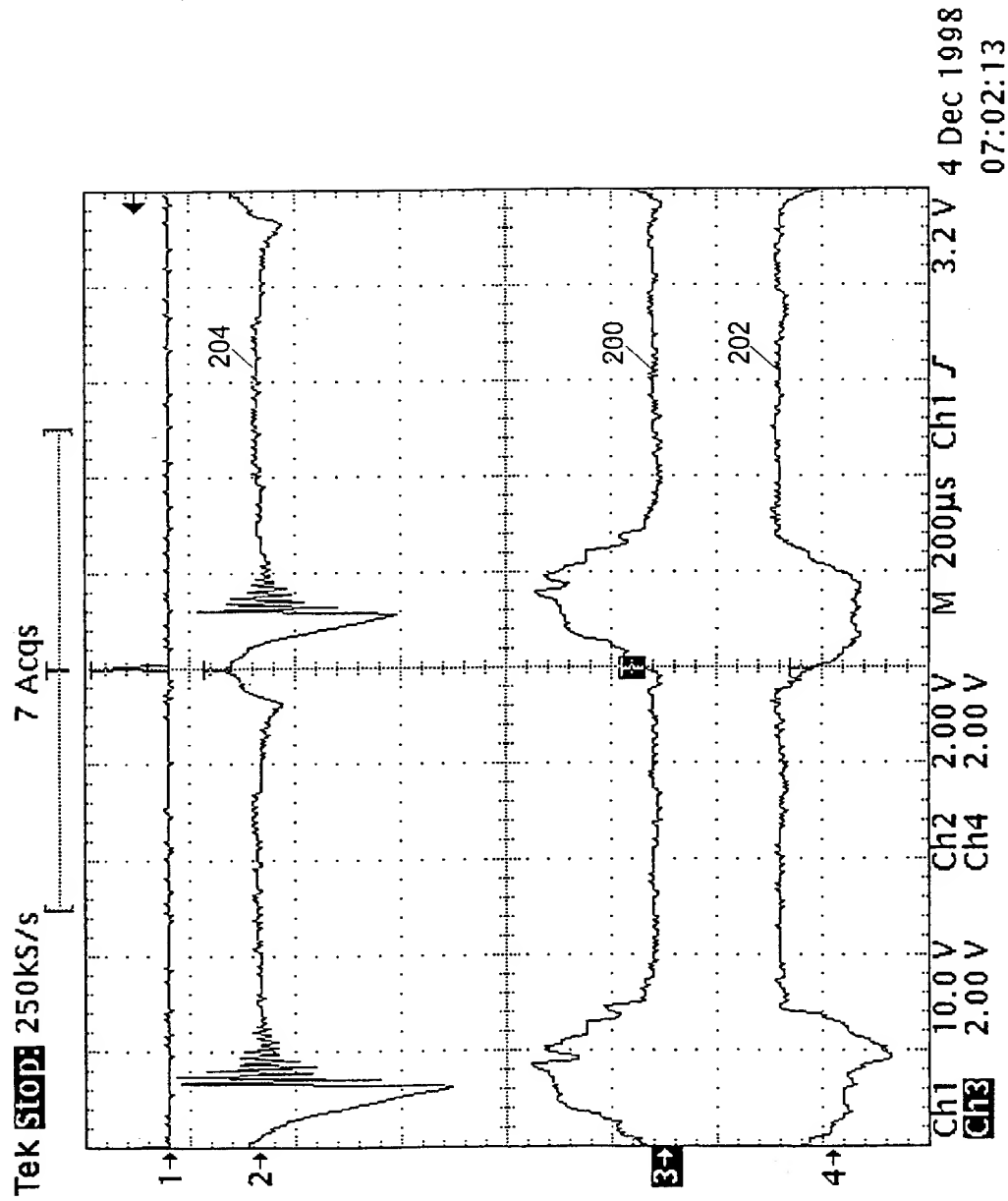


Fig. 11

Polymer sample,  
image size 25  $\mu\text{m}^2$ ,  
93kHz / 1kHz

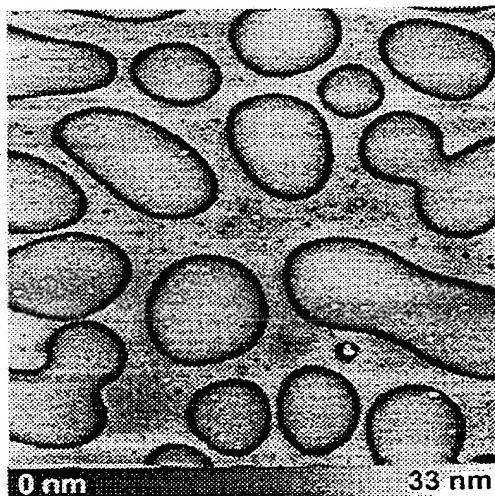


Fig.12A

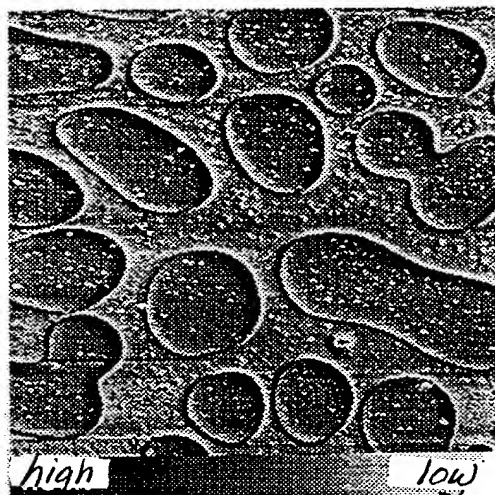


Fig.12B

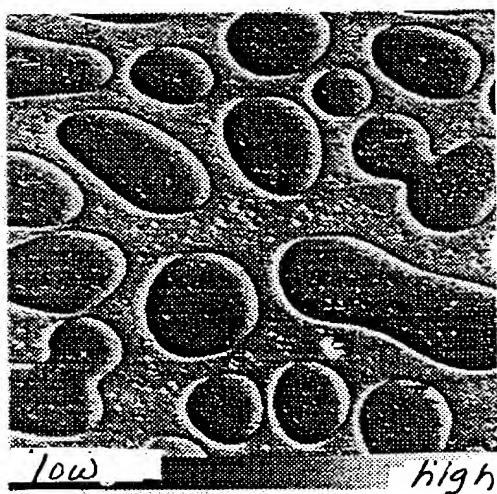


Fig.12C

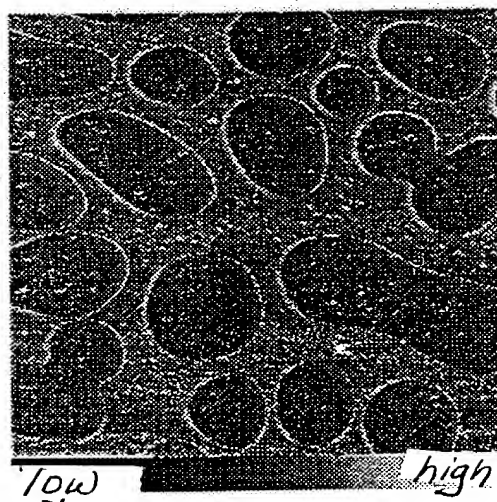


Fig.12D

Polymer sample,  
image size 25  $\mu\text{m}^2$ ,  
230kHz / 1kHz

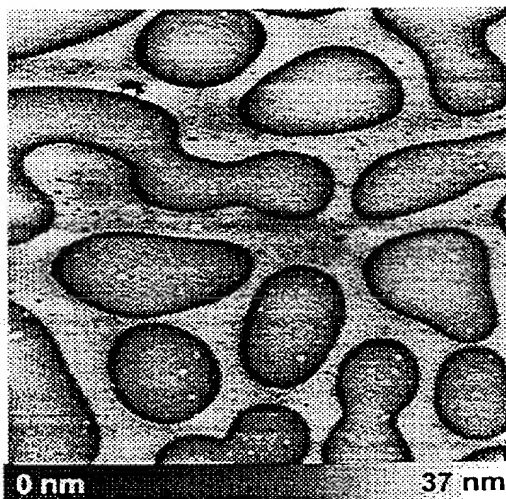


Fig.13A

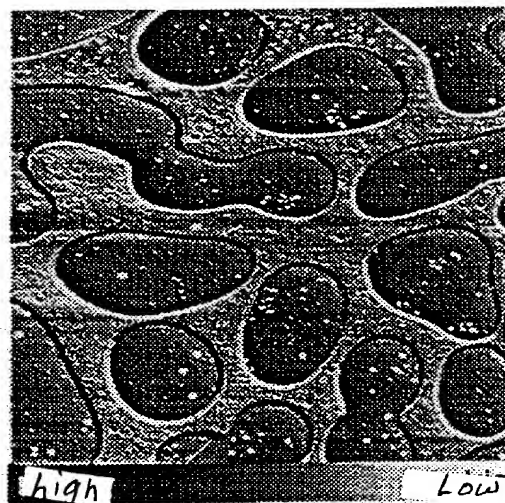


Fig.13B

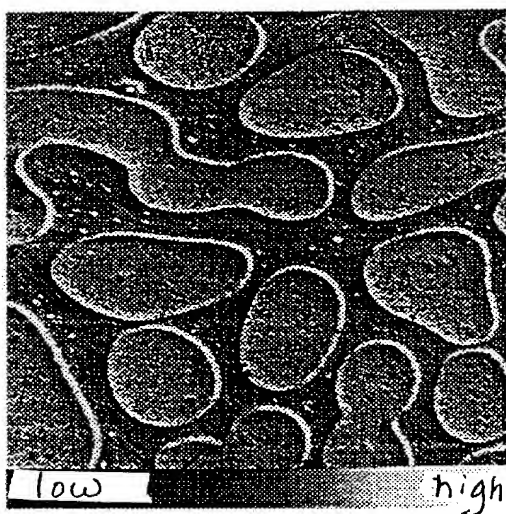


Fig.13C

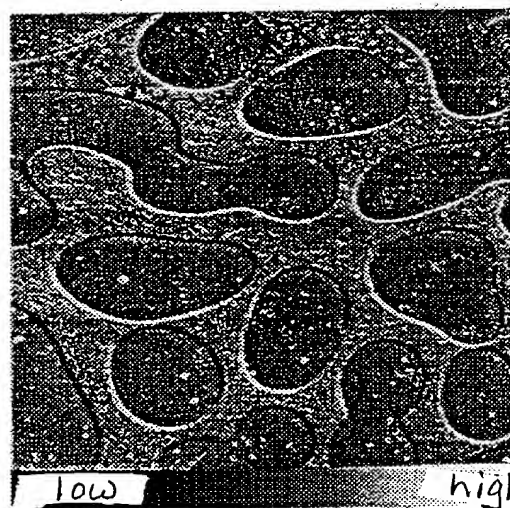


Fig.13D

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T-578 P.03/00 Job-193

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Patent and Trademark Office: U.S. DEPARTMENT OF COMMERCE

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As a below named inventor, I hereby declare that:

My residence, post office address and citizenship are as stated next to my name.

I believe I am the original, first and sole inventor (if only one name is listed below) or an original, first and joint inventor (if plural names are listed below) of the subject matter which is claimed and for which a patent is sought on the invention entitled:

METHOD AND DEVICE FOR SIMULTANEOUSLY  
DETERMINING THE ADHESION, FRICTION, AND OTHER  
MATERIAL PROPERTIES OF A SAMPLE SURFACE

deren Beschreibung hier beigefügt ist, es sei denn (in diesem Falle Zutreffendes hier ankreuzen), diese Erfindung

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Vertrags über die Zusammenarbeit auf dem Gebiet des  
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\_\_\_\_\_ abgeändert (falls  
zutreffend).

☒ was filed on January 4, 2000  
as United States Application Number or PCT  
International Application Number PCT/DE00/00003  
and was amended on \_\_\_\_\_  
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I hereby state that I have reviewed and understand the contents of the above identified specification, including the claims, as amended by any amendment referred to above.

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I acknowledge the duty to disclose information which is material to patentability as defined in Title 37, Code of Federal Regulations, § 1.56.

[Page 1 of 3]

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#### Prior Foreign Applications (Frühere ausländische Anmeldungen)

199 00 114.6 Germany  
 (Number) (Country)  
 (Nummer) (Land)

                                           
 (Number) (Country)  
 (Nummer) (Land)

#### Priority Not Claimed ~~Priorität nicht beansprucht~~

05 January 1999  
 (Day/Month/Year Filed)  
 (Tag/Monat/Jahr der Anmeldung)

                      
 (Day/Month/Year Filed)  
 (Tag/Monat/Jahr der Anmeldung)

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 (Application No.) (Filing Date)  
 (Ankennzeichen) (Anmeldetag)

                                           
 (Application No.) (Filing Date)  
 (Ankennzeichen) (Anmeldetag)

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DE00/00003 January 4, 2000  
 (Application No.) (Filing Date)  
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